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An Improved Decay Scheme for Cesium-134.

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AN IMPROVED DECAY SCHEME FOR Cs^{134}

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ABSTRACT

A study of the radioactive decay of Cs^{134} has been made, using scintillation spectrometer techniques. Previous work with this nucleus has demonstrated that the decay scheme is unusually complex. Nine gamma rays are known, and the beta spectrum has been shown to consist of three or more components. In the present work, coincidences of gamma rays with gamma rays, and of gamma rays with beta rays have been measured. The gamma ray spectrum coincident with each of the major gamma rays has been measured using two 3" by 3" NaI(Tl) crystals. The beta rays coincident with each of the major gamma ray photopeaks in a NaI(Tl) crystal have been measured using an anthracene crystal for the electron detection. The beta ray spectrum has been measured with an anthracene crystal, and has been found to contain beta rays of higher energy than any previously reported. Additional beta ray components have been observed with end point energies of approximately 350, 1000, and 1300 kev. A decay scheme is proposed which accounts for the observed coincidences and which is consistent with the relative intensities of the gamma rays and beta ray components.

CHAPTER I

INTRODUCTION

The determination of the energy levels of atomic nuclei is an important problem of experimental nuclear physics. The prediction of such energy levels, and of the probability of the transitions which take place between them, is an essential task of theoretical nuclear physics, and any successful theory must account for the observed energy levels and transitions. The testing of theories is often limited by inadequate knowledge of the actual energy levels of nuclei.

There are several methods by which energy levels may be measured. Among them are the study of nuclear reactions, scattering of particles by nuclei, Coulomb excitation of the energy levels in nuclei, and by the observation of radiations resulting from radioactive decay. In the last named method, use is made of the fact that in alpha and beta decay, the daughter nucleus is often left in an excited state, from which it decays by gamma ray emission. Measurement of the alpha, beta, and gamma ray energies, and the study of such features as the internal conversion coefficients of the gamma rays, angular correlations between successive radiations, and coincidences between two or more emitted radiations (which shows them to be in sequence) provides much information about the energy levels of the daughter nucleus. Studies of this kind can lead not only to an understanding of the radioactive decay scheme of the nucleus, but coupled with the theory of angular correlations between successive radiations, can often provide spins and parities for the levels which participate in the radioactive decay process.

In the study of beta decay processes, determination of the beta ray energies can be made with a magnetic beta ray spectrometer. The gamma ray energies too can be determined in the magnetic spectrometer by measurement of the internal conversion electron energies, or by measuring the energies of the photoelectrons produced in a high atomic number radiator. Determination of the relative intensities of the gamma rays can be accomplished with a magnetic spectrometer from the intensities of the photoelectrons, produced in a radiator, or more directly by a scintillation spectrometer. Coincidence between successive radiations can be determined in several ways. The most efficient method employs scintillation spectrometer techniques to select the particular radiations between which coincidences are to be investigated.

The nucleus Cs^{134} is one whose decay scheme is imperfectly understood. This nucleus is known to decay by negative beta emission to excited states of the daughter nucleus Ba^{134}. The complexity of the beta and gamma ray spectra indicates that a number of levels are involved. It is therefore profitable to study the decay in order to determine the energy levels, and to understand the radioactive decay scheme.

The earliest intensive investigation of Cs^{134} was made by Elliot and Bell.[1] Since that time, a number of studies have been made of the beta and gamma ray spectra. Some of these studies have included gamma-gamma and beta-gamma coincidences. The decay of Cs^{134} has been shown to involve at least eleven gamma rays and four, five or more beta ray components, Several different decay schemes have been proposed. A review of the work done in the study of Cs^{134} has been given by Fayard.[2]

The gamma rays from Cs^{134} have been measured in magnetic spectrometers by Waggoner, Moon, and Roberts,[3] Joshi and Theosar,[4] Bertolini,[5] Cork *et al.*,[6] Keister, Lee and Schmidt,[7,8] and by August.[9] Gamma rays have been reported with energies of, or in the neighborhood of 202, 473, 563, 569, 605, 796, 801, 1038, 1168, 1367, and 1401 kev. The numbers assigned are

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[1] L. G. Elliot and R. E. Bell, Phys. Rev. 72, 969 (1947).

[2] O. E. Fayard, Thesis, Louisiana State University (1957).

[3] M. A. Waggoner, M. L. Moon, and A. Roberts, Phys. Rev. 80, 420 (1950).

[4] M. C. Joshi and B. V. Theosar, Phys. Rev. 96, 1022 (1954).

[5] G. Bertolini, Nuevo Cim. 2, 273, 746 (1955).

[6] J. M. Cork, J. M. Leblanc, W. H. Nester, D. W. Martin, and M. K. Brice, Phys. Rev. 90, 444 (1953).

[7] F. H. Schmidt and G. L. Keister, Phys. Rev. 86, 632 (1952).

[8] G. L. Keister, E. B. Lee and F. H. Schmidt, Phys. Rev. 97, 451 (1955).

[9] L. August, Dissertation, Louisiana State University (1957).

those due to Keister, Lee and Schmidt, except for the 202 and 1401 kev gamma rays, which they did not observe.

Scintillation spectrometer studies have been made by Bertolini,[5] Chandra,[10] and Fayard[2] to determine coincidences between gamma rays, and by Gabro[11] to determine relative intensities of the gamma rays.

Cork, et al.[6] have studied the beta spectrum, using a magnetic spectrometer. They have resolved the beta spectrum into four components at 80, 210, 410 and 657 kev. Keister, Lee and Schmidt[8] have found components at 83, 308, 655, and 683 kev. They report that their data might allow the component reported at 308 kev to be resolved into two components.

Keister, Lee, and Schmidt[8] have measured the relative intensities and conversion coefficients for the major gamma rays. They have assigned multipolarities to the gamma rays on the basis of these measurements.

The coincidences between gamma rays previously reported have been tabulated by Fayard,[2] along with those which he observed. They are reproduced in Table I.

The recently proposed decay scheme of Keister, Lee and Schmidt,[8] shown in Figure 1, has made use of more experimental material than any previous scheme. Certain features, such as the cascade of the 569, 796, and 605 kev gamma rays, seem to be on a firm footing. Other features are less convincing.

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[10] G. Chandra, Proc. Ind. Acad. Sci., 44A, 4, 194 (1956).

[11] A. N. Gabro, Thesis, Louisiana State University (1956).

TABLE I

COINCIDENCES OBSERVED BY OTHERS

	Coincidences
Deutsch and Siegbahn	568-602-799
Bertolini	560-600, 600-800, 600-1350
Chandra	460-604, 555-604 604-794, 604-1399
Fayard	563-600 600-800 800-800 800-1168 800-1368 200-1368 800-920 (some indication 1038-1368 ?)
	Non Coincidences
Chandra	460-794, 794-801, 794-1349
Fayard	600-1168, 600-1038

The scheme is a modification of one previously proposed by Cork *et al.*[6] The changes in the scheme were made largely to accord with the multipolarities which they assigned to the gamma rays, and with their partial investigation of coincidences between the beta rays and some of the gamma rays.

A thorough investigation of the coincidences among all the known gamma rays and between the gamma rays and the electron spectra was proposed, in the hope that it would allow a more satisfactory decay scheme to be developed. Because of the large number of radiations present in both the gamma and the beta ray spectra, there are a great number of coincidences to be investigated. Because of the inefficient geometry

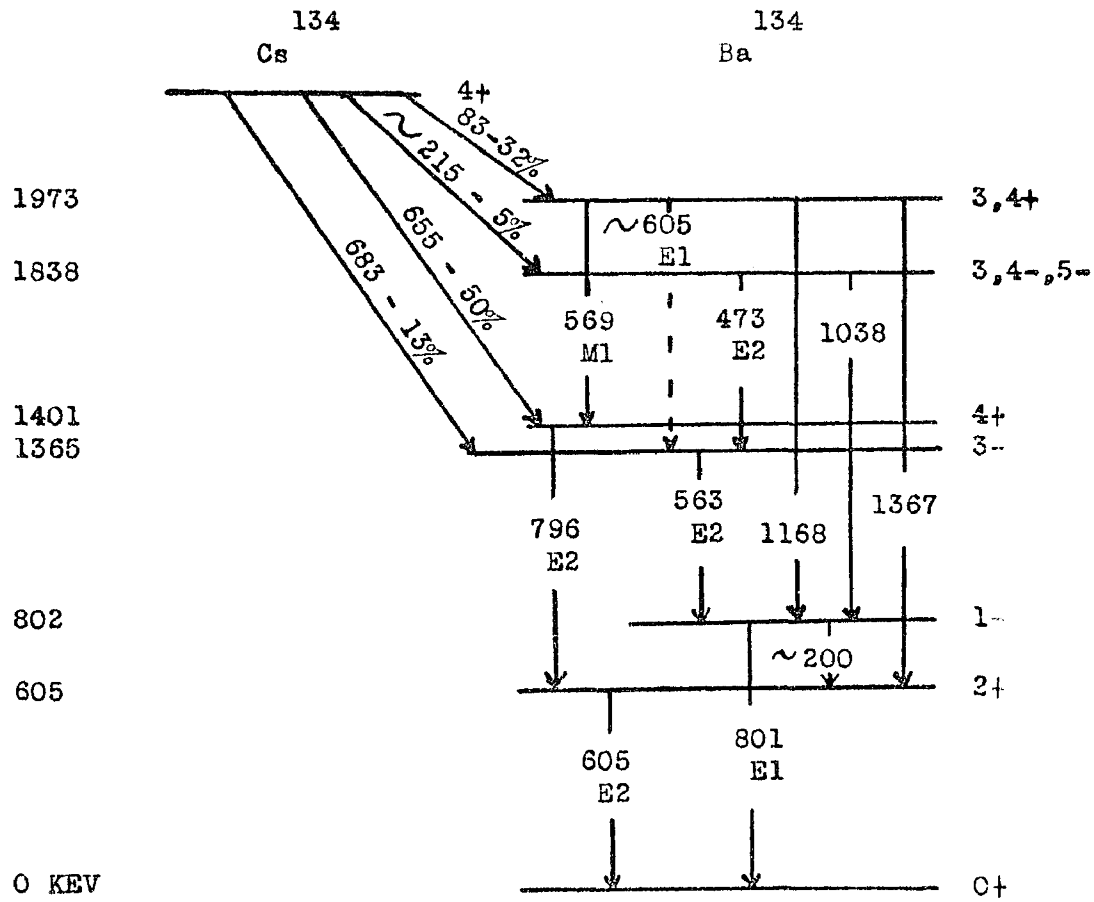

Figure 1. Decay scheme for the ground state transitions proposed by Keister, Lee, and Schmidt (reference 8).

possible when using two NaI(Tl) crystals as detectors, low counting rates are observed. Using a twenty channel analyzer, however, such a coincidence counting program can be accomplished in a reasonable length of time.

Such a program was undertaken using the apparatus of Figure 2. The single channel analyzer was held fixed at a pulse height corresponding to each of the major gamma ray photopeaks, while the twenty channel analyzer scanned the spectrum of gamma rays, or beta rays in the beta-gamma experiment, in coincidence with the pulses from the single channel analyzer.

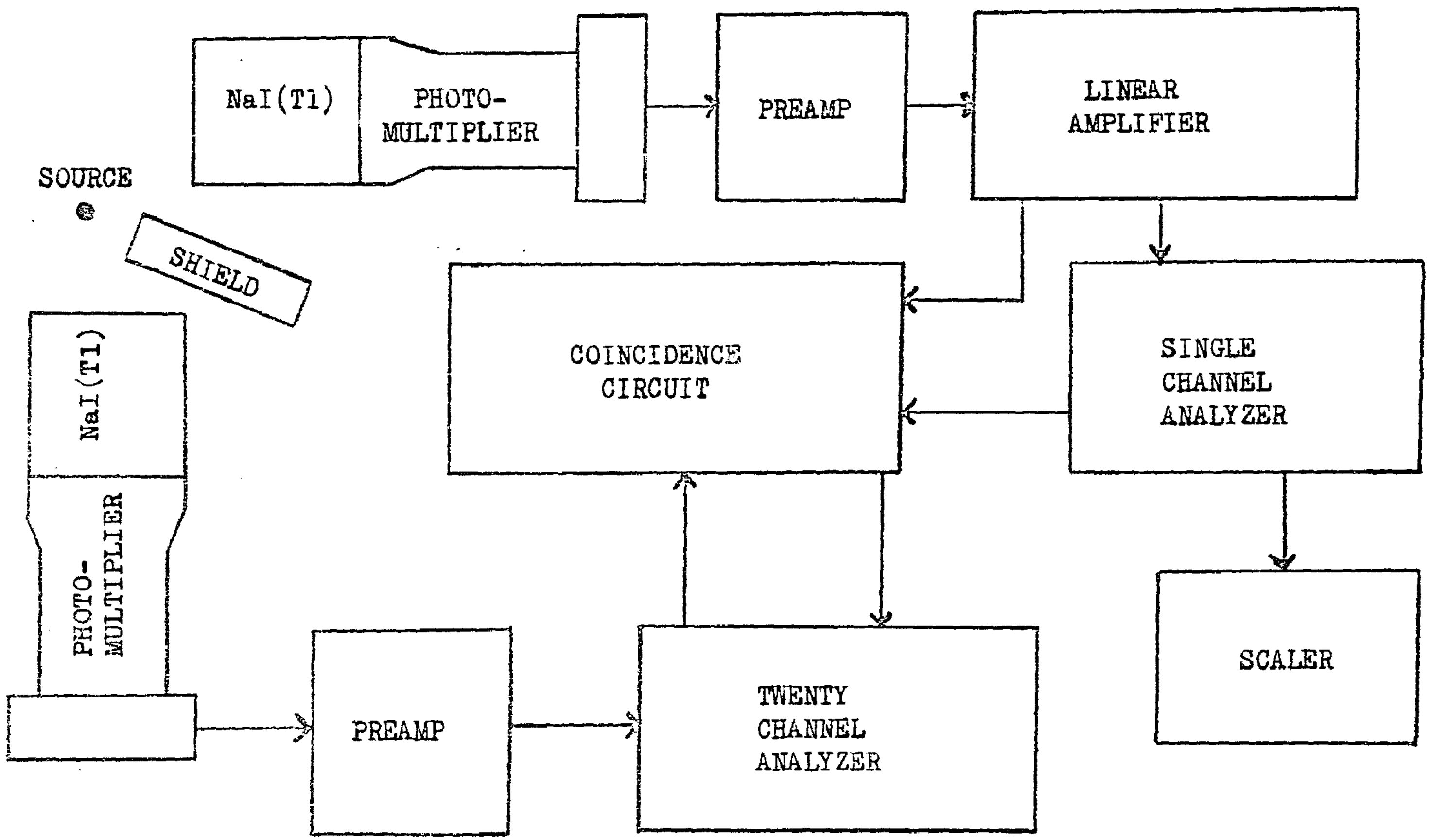

Figure 2. Block diagram of the equipment

CHAPTER II

EXPERIMENTAL PROCEDURES

The Cs^{134} sources were prepared from a CsCl solution in HCl obtained from the Oak Ridge National Laboratory. The Cs^{134} was made by neutron capture in Cs^{133} in a pile. For the gamma-gamma coincidence study, a source of about two microcuries was prepared by evaporation on an aluminum foil backing. For the beta-gamma coincidence study, a source of about one microcurie strength was prepared by evaporation on a thin (2.7 mg/cm^2) plastic backing. The latter source was left uncovered to reduce absorption of the beta rays. Because of the thin backing, scattering by the source backing was small. The principal absorption was that in the 3 cm of air between the source and the crystal. The source thickness was of the order of 1 mg $/cm^2$, and scattering in the source was small.

Two 3" x 3" NaI(Tl) crystals were used for the gamma-gamma coincidence counting. These were mounted on Dumont 6363 photomultiplier tubes. For the beta-gamma counting, one of the NaI(Tl) crystals was replaced by a 1 1/2" x 1/4" anthracene crystal. This was mounted on a Dumont 6292 photomultiplier tube.

The apparatus used in the coincidence counting observations is shown in block diagram in Figure 2. The arrangement for the gamma-gamma coincidences and for the beta-gamma coincidences was the same, with the exceptions that the crystal used for the electrons was anthracene instead of sodium iodide and that the graded shield was removed to make room for an aluminum tube to cover the crystal and photomultiplier for the electron detection. The source in this case was inside the aluminum tube. The tube was necessary to exclude light from the photomultiplier, since the crystal was covered by a very thin aluminum foil which had pinholes in it.

The pulses from one of the phototubes were amplified and then analyzed by a single channel analyzer. Provision was made at this point to count the pulses from the single channel analyzer so that the pulse spectrum from that photomultiplier could be determined.

The pulses from the other phototube were fed to a twenty channel pulse height analyzer. A fast-slow coincidence circuit was employed to gate the twenty channel analyzer. A gate pulse was obtained only if there were pulses coincident in the two photomultipliers within 0.2 microseconds as well as a pulse of appropriate size in the single channel analyzer. Only then would the twenty channel analyzer register a pulse, in its appropriate channel.

The speed of the coincidence circuit was more than adequate for the coincidence investigation. Since Keister, Lee, and Schmidt[8] have shown that there are no delayed coincidences with a half life greater than 0.2 microseconds, the coincidences circuit was not suitable for use in a search for delayed coincidences.

The coincidence counting rates per channel in the twenty channel analyzer varied in the different runs from the order of 0.001 to 1 count per second. For the major portion of the work, the gamma ray spectrum was spread out over a 100 channel interval of the 120 available channels in order not to limit the resolution by that of the analyzer. Much of the data required long time intervals to collect a statistically significant number of counts. During these time intervals, small drifts in the gain of the system were observed. The possibility of a large drift in the system was guarded against by frequent checks on the gain during operation.

The coincidence circuit is essentially that described by Fayard[2] except for the minor modification needed to produce a gating pulse for the twenty channel analyzer. Appropriate delays selected from experimentally determined curves of instrumental delay versus pulse height were inserted to compensate for pulse delays in the circuit. In this way it was possible to ensure that two pulses which were actually in coincidence would be counted in the twenty channel analyzer over the entire twenty channel range. The possibility that pulses delayed in the source by less than 0.2 microsecond may have caused a loss of counting efficiency near the ends of the twenty channel spread must be considered. However, the experimental curves of instrumental delay versus pulse height indicated that this effect was small and moreover very few of the photopeaks fell near the ends of the twenty channel spread.

The single channel and twenty channel energy scales for the gamma rays were determined from the photopeaks for the major gamma rays, using the measurements of Keister, Lee and Schmidt[8] for the energies. The energy scale in the anthracene crystal for the electron spectra was determined from the conversion electron peaks seen in this spectrum. Figure 3 shows the twenty channel energy scale. This scale applies to all the gamma ray spectra shown in the illustrations.

In the gamma-gamma coincidence counting, a 1/4" lucite shield was placed over each crystal to exclude electrons. In the beta-gamma coincidence counting, the NaI(Tl) crystal detecting the gamma rays was shielded from electrons. However, the anthracene crystal was not shielded from the photons. Therefore, it was necessary to make measurements of the gamma-ray intensity in the anthracene crystal, by placing an electron shield over the crystal. This was then subtracted from the first observed spectrum to give the electron spectrum.

The beta spectrum energies were corrected for air absorption in all cases. The absorption in the thin aluminum foil reflector covering the anthracene crystal was negligible. No attempt was made to collimate the beta rays. This resulted in more scattering out of the crystal than with a collimated source, but the requirements in terms of counting rates made it desirable to have the crystal subtend a larger solid angle than would have been possible with a collimated source.

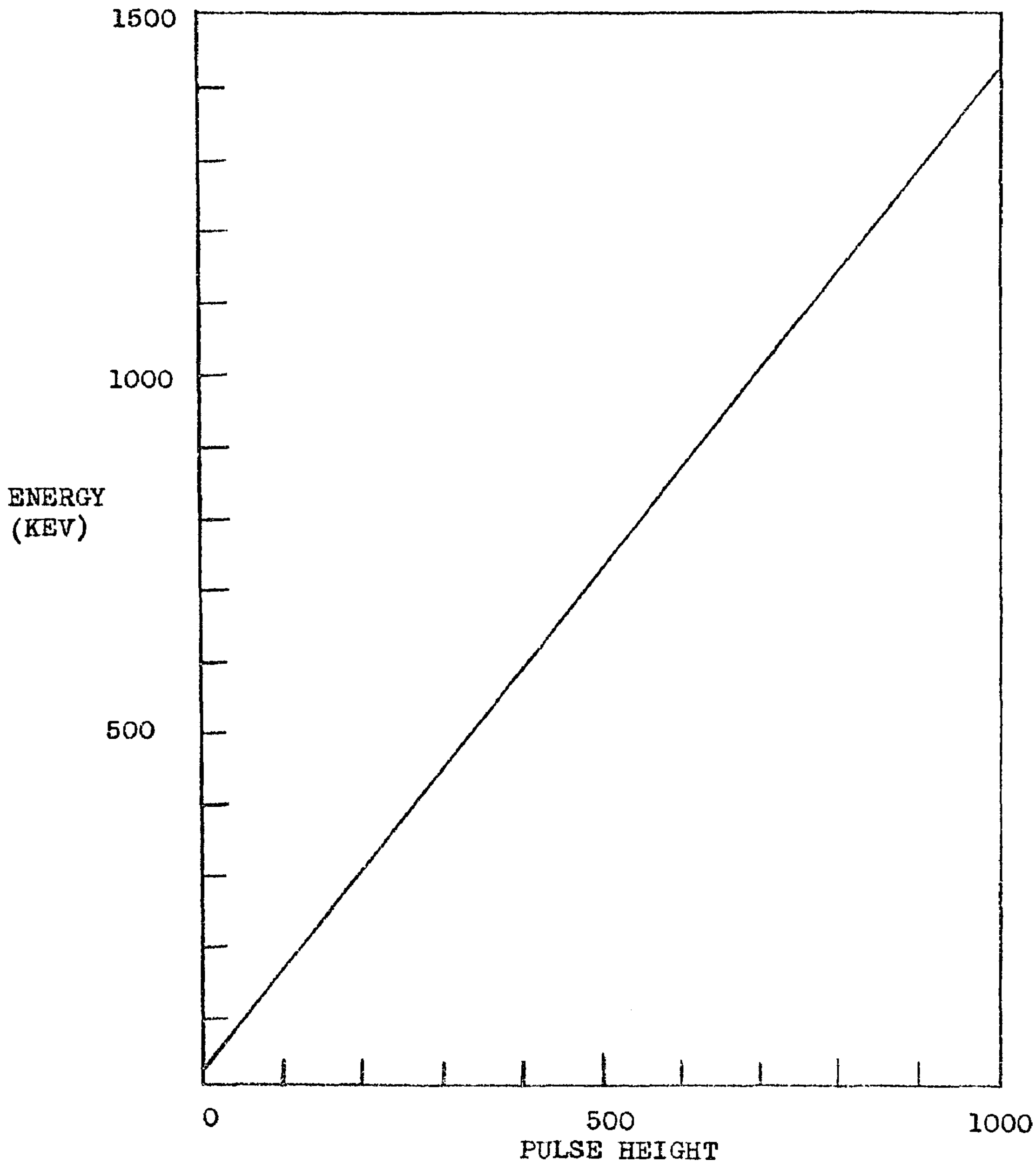

Figure 3. Energy scale for the gamma ray spectra.

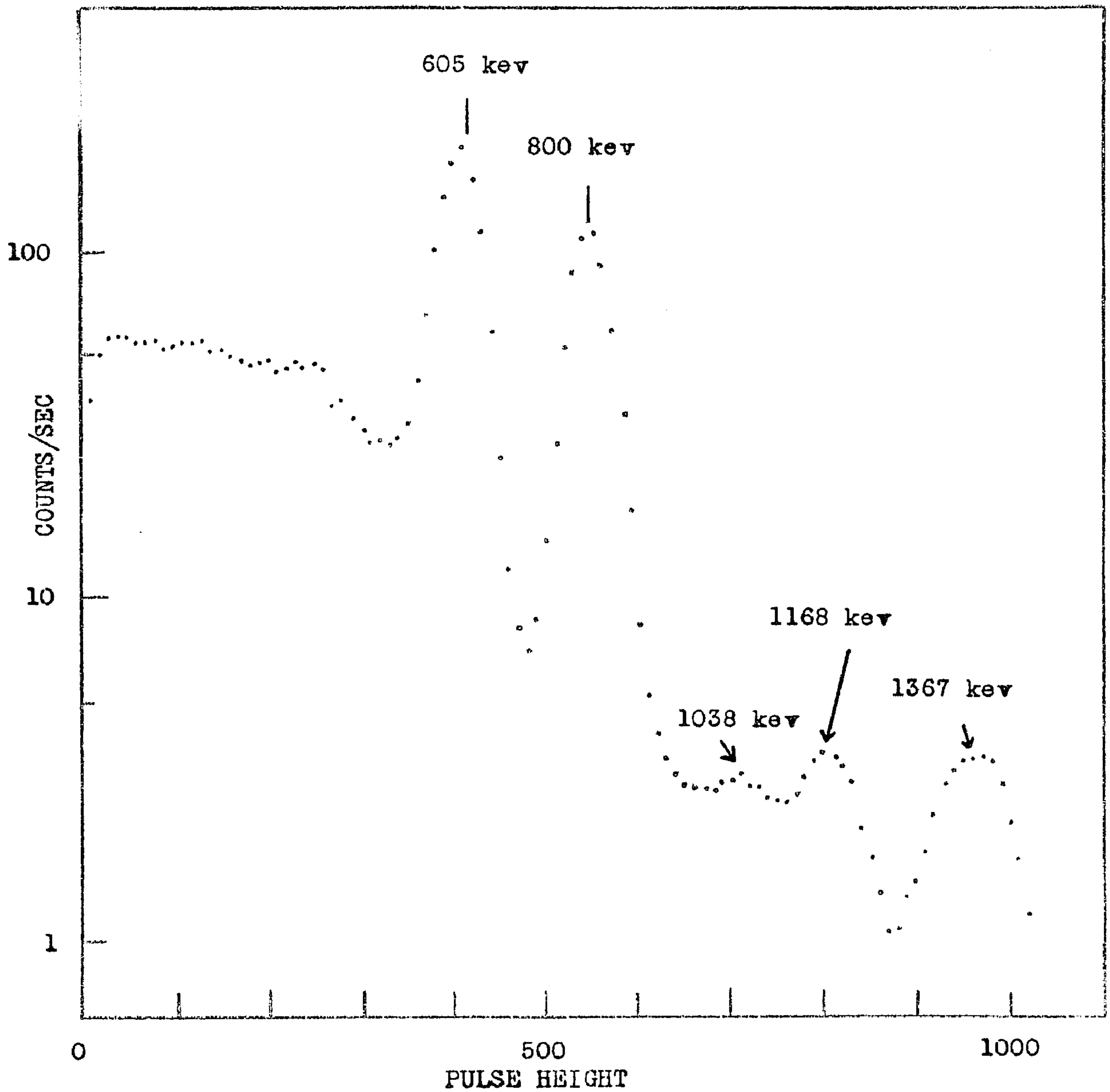

Figure 4. Gamma ray singles spectrum.

CHAPTER III

OBSERVED COINCIDENCES

In the gamma-gamma coincidence counting program, coincident photopeaks were observed for all the known gamma rays of Cs^{134} which could be resolved with a NaI(Tl) crystal. In addition, by using an expanded scale, the peaks near 600 kev could often be shown to be due to more than one gamma ray. In each of these cases, the observed peak was resolved into two peaks by fitting a gaussian distribution of appropriate width to the high energy side of the curve and subtracting this from the observed peak. In each case there was left a gaussian peak of the same width at such an energy that, if the higher energy peak be identified with the 605 kev gamma ray, the other peak could be either the 563 or 569 kev photopeak, or both.

Photopeaks for a gamma ray of the order of 280 kev were observed in several coincidence spectra. There is a doubtful case of a gamma ray of 130 kev energy in coincidence with the 1038 kev gamma ray.

In Table II the gamma-gamma coincidences observed in this study are shown in the first two columns. The numbers in column 2 are the percentages of the gamma rays of column 1 which are in coincidence with the gamma rays of column 2. The other columns of the table show a summary of the results of

Keister, Lee, and Schmidt,[8] using a magnetic spectrometer.

TABLE II

GAMMA-GAMMA COINCIDENCES OBSERVED IN THIS WORK

Gamma ray (kev)	Gamma-gamma coincidences observed	Multipole order assigned by Keister, *et al*.	Relative intensity, Keister, *et al*.
280	473, 1038, 1168, 1367		
473	280, 605, 800, 1168, 1367	E2	1.8 ± 0.5
563 569	605, 800, 1168, 1367	E2 M1	9.4 ± 2 12.8 ± 2
605	473 (4.5%), 563-569 (25%), 605 (25%), 800 (83%), 1040 (1%), 1367 (1%)	E2	100
796 802	605 (100%), 563-569 (40%), 1168 (1.5%)	E2 E1	91 ± 4 18 ± 4
1038	127 (8%), 280 (16%), 473 (17%), 563-569 (35%), 605 (70%), 800 (20%)	E3 or M1	0.9 ± 0.2
1168	280 (11%), 473 (14%), 563-569 (25%), 605 (18%), 800 (49%)	E1 or E2	3 ± 0.4
1367	280 (1%), 563-569 (27%), 605 (56%)	E1 or E2	4.6 ± 0.3

The coincidence fractions recorded were calculated from the relation

$$f = \frac{C}{N e_p F \Omega},$$

where f is the fraction of the gamma decays of the particular gamma ray in the first column which is coincident with the gamma ray in the second column, C is the observed coincidence counting rate at the photopeak for the second column gamma ray (in the twenty channel analyzer), N is the counting rate for the first column gamma ray (in the single channel), e_p is the intrinsic peak efficiency of the twenty channel crystal, F is the fraction of the photopeak included in the peak counting channel of the twenty channel analyzer, and Ω is the fraction of 4π solid angle subtended by the twenty channel analyzer's crystal at the source. The fractions so calculated are expressed as percentages in the table.

Except for the 280 kev gamma ray, each case was studied by determining the gamma ray spectrum, in the twenty channel analyzer, which was coincident with the gamma ray in question in the single channel analyzer. In the case of the 280 kev gamma ray, the coincidences noted in the first column were observed with the single channel analyzer set to count pulses corresponding to the energies in column 2.

Taking each gamma ray in turn, the following comments are appropriate. In each case where a 280 kev was observed, the identification depended on the result of subtracting a constructed Compton distribution for the 473 kev, 600 kev, and 800 kev gamma rays, which are in total in each case of approximately the same intensity as the peak itself. The fraction of coincidences calculated is in this case subject to more than the usual 5 to 10% error, and is probably correct only as to order of magnitude.

The spectrum of gamma rays coincident with the 473 kev gamma ray, (Fig. 5), has been corrected for coincidences due to Compton electrons produced in the crystal by the 600 and 800 kev gamma rays. This correction was accomplished by

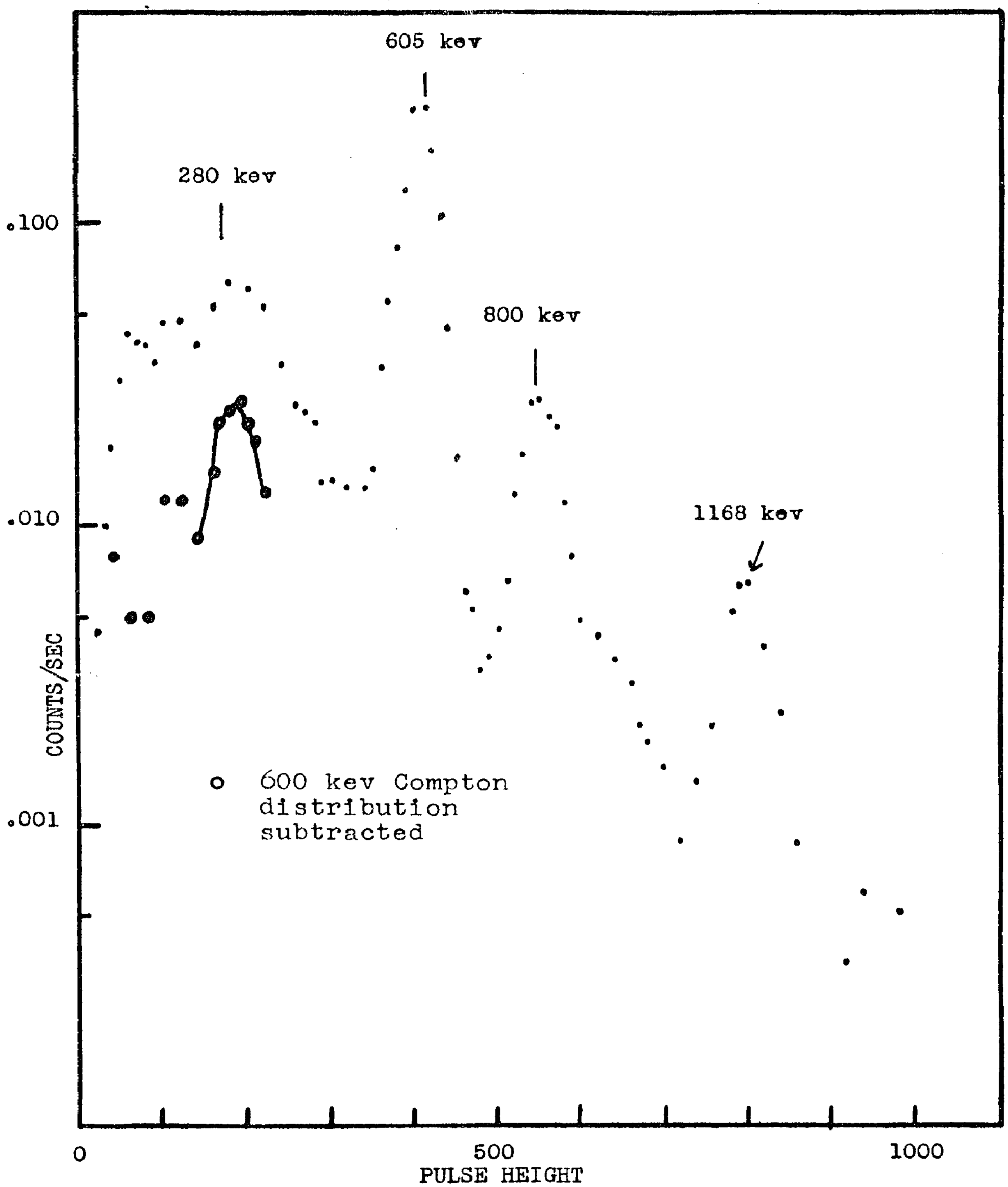

Figure 5. Gamma ray spectrum coincident with the 473 kev gamma ray.

subtracting 13% of the spectrum coincident with the 800 kev gamma ray peak, and 4% of the spectrum coincident with the 600 kev gamma ray peak. A further complication arises from the fact that the 473 kev photopeak is not visible in the singles spectrum due to its low intensity. This makes it impossible to assign a number for the single channel counting rate for the 473 kev gamma ray with any certainty. For this reason, coincidence fractions have not been given.

The 563, 569, and 605 kev gamma rays can not be resolved by the crystal. The gamma ray spectrum observed in coincidence with the peak corresponding to 600 kev then contains elements in coincidence with each of the 563, 569, and 605 kev gamma rays. The spectrum coincident with the 600 kev peak is shown in Figure 6. Comparison with the singles spectrum (Fig. 4) shows that there is a peak at about 600 kev, with a significant shift toward lower energies of this peak in the coincidence spectrum. This peak can be resolved into two peaks by subtracting a gaussian distribution of appropriate width for 605 kev, fitted to the high energy side of the observed peak. The peaks are of approximately equal height, and the lower energy peak corresponds to a gamma ray of about 565 kev, which may be due to the 563 kev, the 569 kev gamma ray or both, while the higher energy peak corresponds to 605 kev.

The coincidences listed in column 2 for the 563 and 569 kev gamma rays in column 1 are recorded without fractions. This is because they were deduced from the twenty channel spectra, and not determined by holding the single channel fixed at those energies.

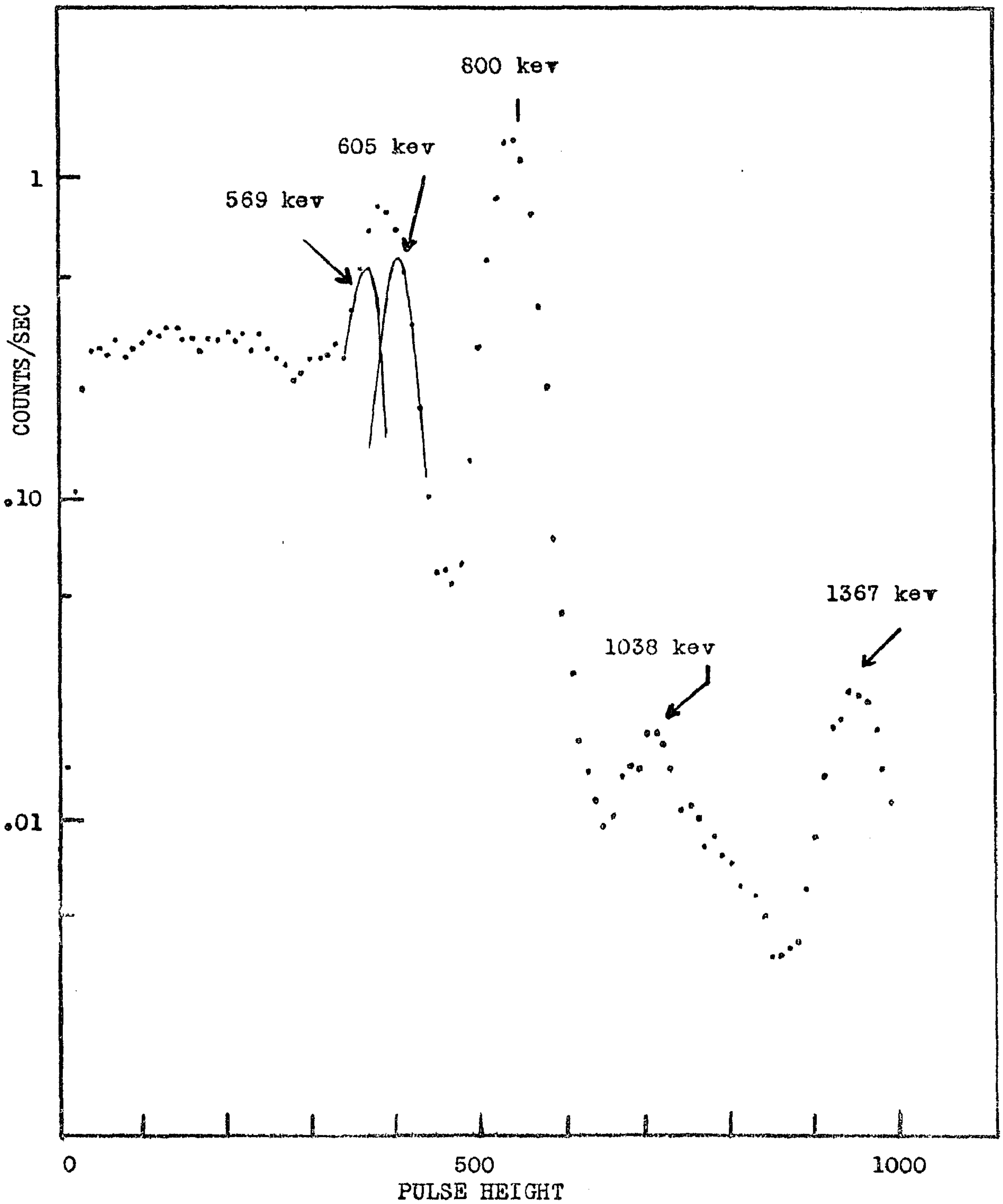

Figure 6. Gamma ray spectrum coincident with the 600 kev gamma rays.

The 796 and 802 kev gamma rays could not be resolved. The coincidences observed (Fig. 7) may be due to either or both. It will become apparent, for instance, that the 1168 kev peak observed in the coincidence distribution is probably due to coincidences with the 802 kev gamma ray, while the coincidences with the 605 kev gamma ray are probably due to the 796 kev gamma ray.

The interpretation of the gamma ray spectrum in coincidence with the 1038 kev gamma ray (Fig. 8) requires that 1/3 of the gamma ray spectrum coincident with the 1367 kev gamma ray be subtracted from the observed distribution, since pulses of the height of the 1038 kev photopeak are composed in part of Compton electrons from the 1367 kev gamma ray. Pulses due to the 1168 kev gamma ray will contribute a much smaller amount to the 1038 kev coincidence spectrum, but an indeterminate amount, since the valley between the 1168 kev photopeak and its Compton distribution falls near 1038 kev, and the actual counting rate is there somewhat indeterminate. This part of the 1168 kev spectrum is visible in the 800 kev coincidence spectrum, and the valley is seen to be 1/10 of the photopeak. No attempt was made to subtract from the 1038 kev spectrum pulses due to the 1168 kev gamma ray but as much as 1/2 of the 800 kev peak observed in coincidence may be due to the 1168 kev gamma ray. The effect on the other peaks is negligible.

The gamma ray spectrum coincident with the 1168 kev gamma ray (Fig. 9), may have as a contribution from the 1367 kev gamma ray as much as 1/6 of the 1367 kev coincidence

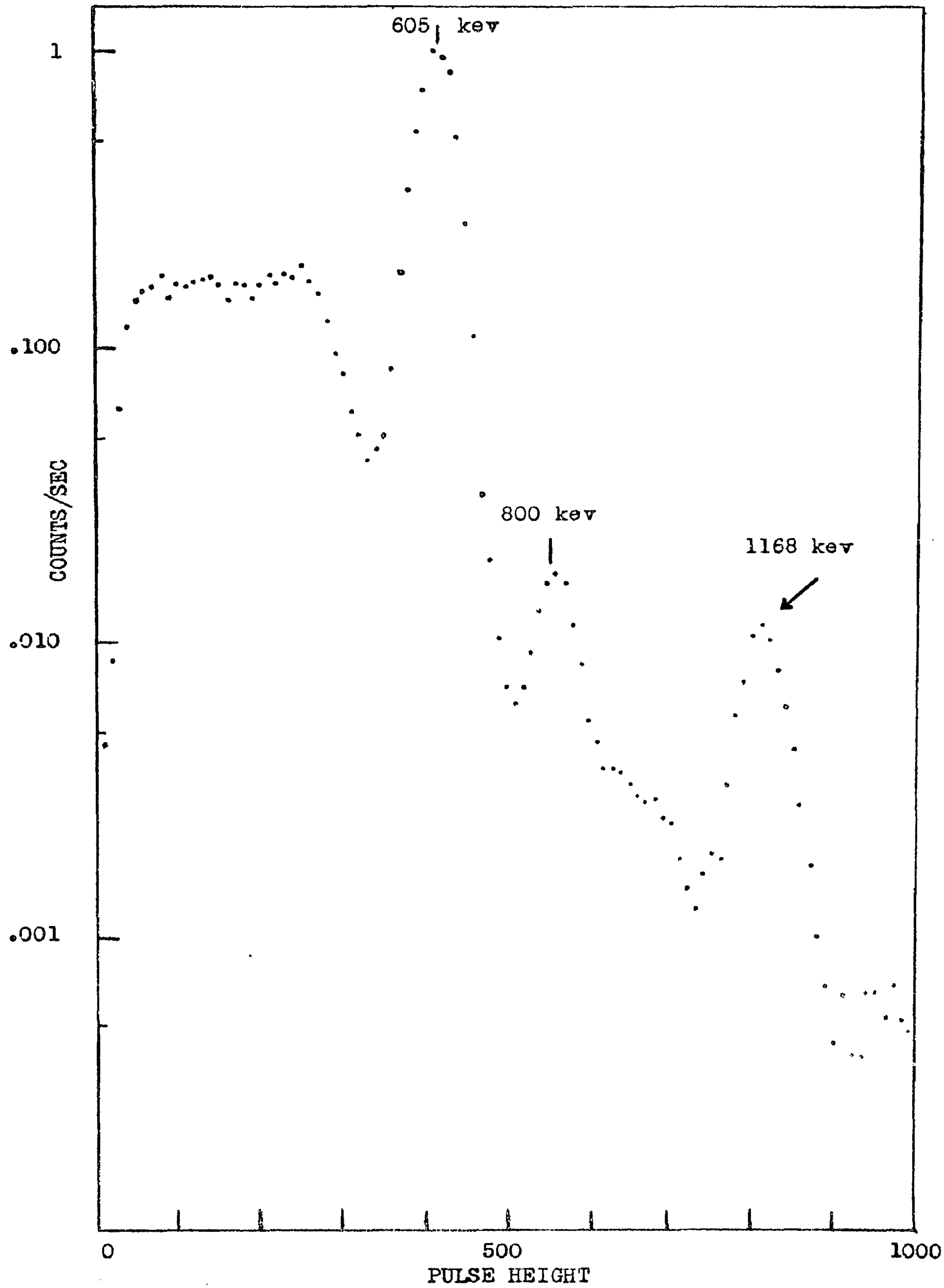

Figure 7. Gamma ray spectrum coincident with the 800 kev gamma rays.

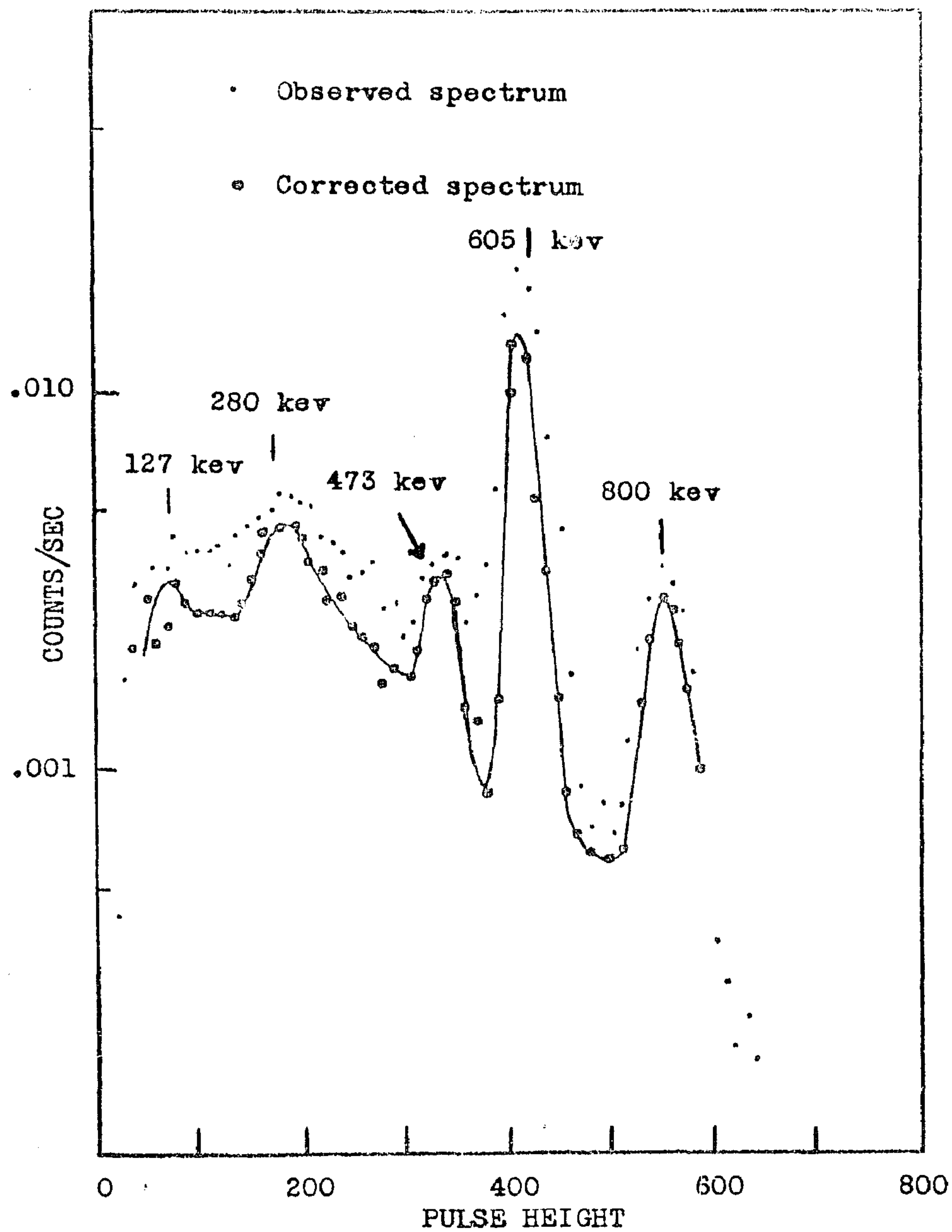

Figure 8. Gamma ray spectrum coincident with the 1038 kev gamma ray.

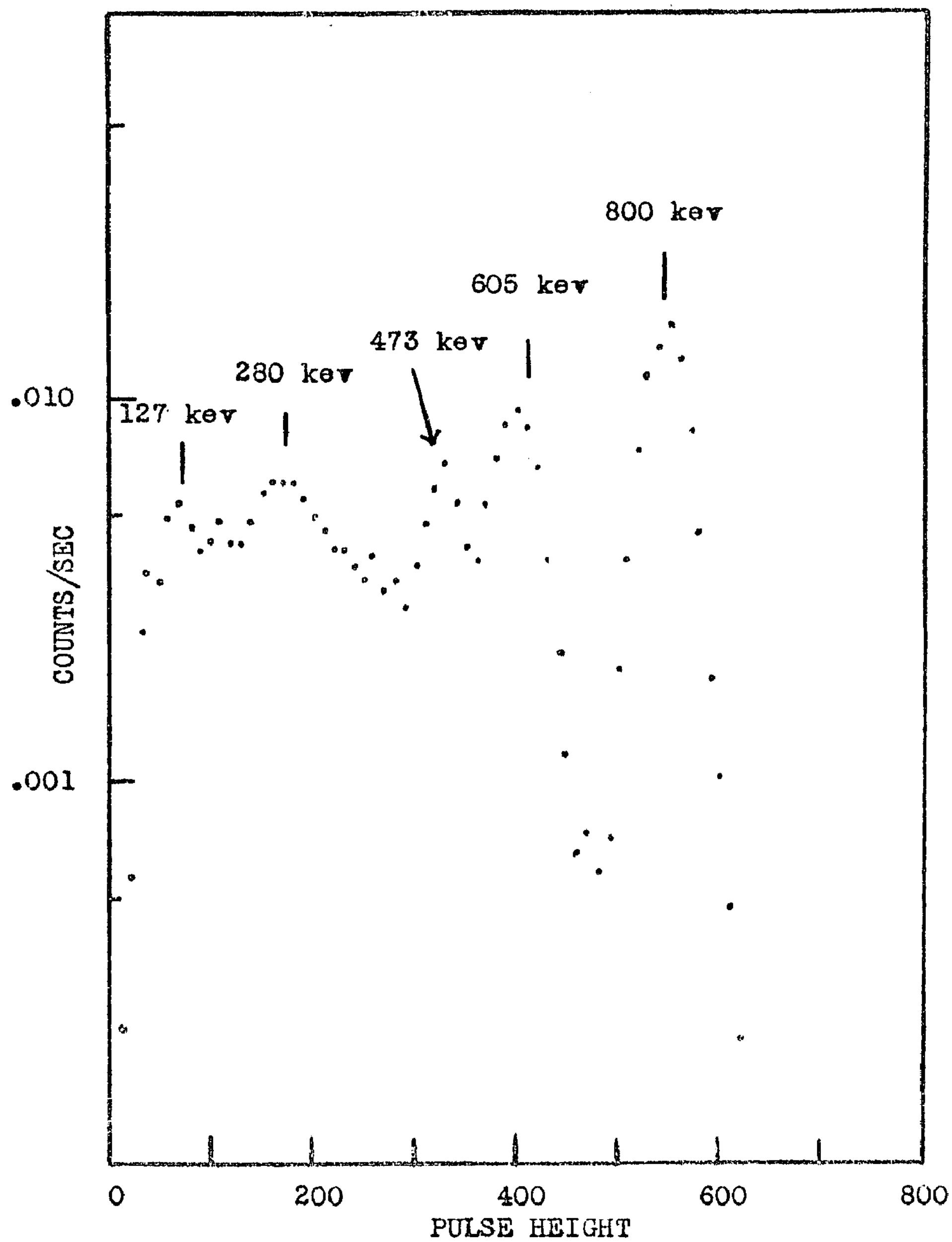

Figure 9. Gamma ray spectrum coincident with the 1168 kev gamma ray.

distribution, but this is a negligible contribution to the observed counting rate.

The gamma ray spectrum in coincidence with the 1367 kev gamma ray (Fig. 10) has no contribution from any other gamma ray, except for sum pulses, since it is the highest energy gamma observed. A weak peak at 800 kev was observed in coincidence with the 1367 kev gamma ray. This peak was about 4 times the random counting rate, determined from

$$R = 2N_1N_2\tau,$$

where R is the random counting rate, N_1 is the counting rate in the single channel, N_2 is the counting rate in the appropriate channel of the twenty channel analyzer, and τ is the resolving time of the coincidence circuit, 2×10^{-7} sec. This gives R = 0.00028. The remainder may be accounted for on the assumption that some of the counts in the 1367 kev peak are due to 800-600 sum pulses, of 1400 kev. The number of these sum pulses can be determined from

$$S = 2N_1N_2\tau,$$

where S is sum pulse rate, N_1 is the counting rate in the single channel at 600 kev, N_2 is the counting rate in the single channel at 800 kev, and τ is the resolving time of the single channel system, 10^{-6} sec. The rate S = 0.28 counts per second. For each of these accidental sum pairs, the 605 gamma ray of the pair will be in coincidence with an 800 kev gamma ray 83% of the time, according to the coincidence data of Table II, or for 0.24 counts per second of the total. Of these, a

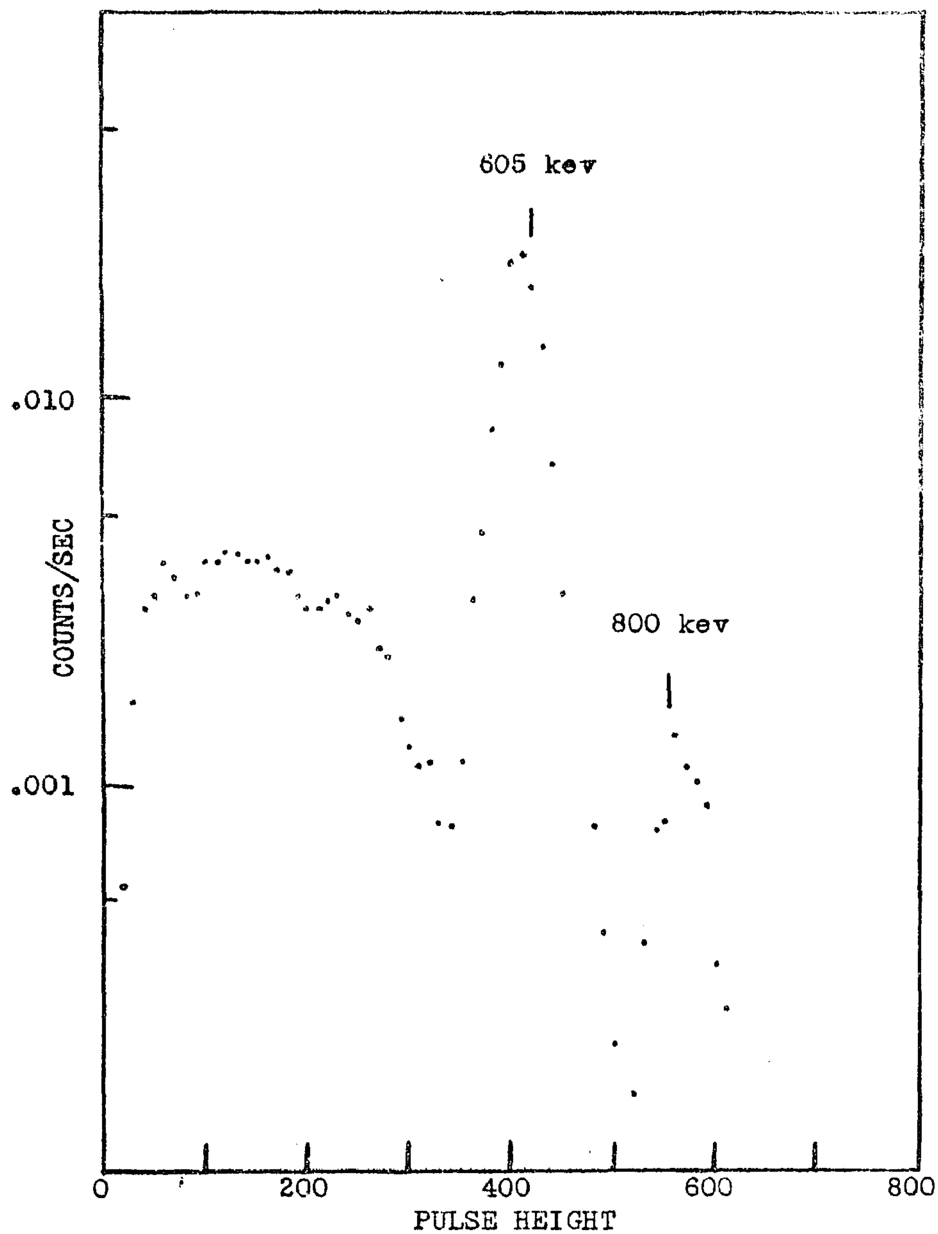

Figure 10. Gamma ray spectrum coincident with the 1367 kev gamma ray.

fraction will be detected in the second crystal, given by

$$f = e_p F \Omega ,$$

where e_p is the intrinsic peak efficiency of the crystal at 800 kev, F is the fraction of the total counts recorded which fall in the peak counting channel, and Ω is the fraction of 4π solid angle subtended by the twenty channel crystal. This gives f = 0.0032. Then, of the counts in the observed 800 peak in the spectrum coincident with the 1367 kev peak, a number given by

$$fS = 0.00077$$

is derived from random sum pulses. The sum of 0.00077 and 0.00028 is 0.001, sufficiently close to the observed counting rate to warrant the assumption that all the observed 1367-800 coincidences are due to this secondary process. It is also significant that in the spectrum coincident with the 800 gamma ray there was no 1367 kev peak observed.

Coincidences between each of the major gamma rays and the beta spectrum were studied, using an anthracene crystal to detect the electrons. In this case, the beta spectrum was analyzed by the twenty channel analyzer. The resolution of the crystal was such that the width of the conversion electron peaks made it difficult to interpret some of the Fermi plots.

The beta-gamma coincidence data was obtained by holding the single channel fixed on each of the following photopeaks: 473, 600, 800, 1038, 1168, and 1367 kev. No electrons below 90 kev were detected, because of the position of the zero on the twenty channel analyzer scale.

The interpretation of the beta spectrum coincident with the 473 kev gamma ray required that 13% of the beta spectrum coincident with the 800 kev gamma rays and 4% of the spectrum coincident with the 600 kev gamma rays be subtracted. When this is done, virtually all of the coincident electrons are accounted for. This suggests that the 473 kev gamma ray may be coincident with the 83 kev beta ray found by Cork _et al._[6] and Keister, Lee, and Schmidt[8] but because of the uncertainties involved in determining the fractions to be subtracted, it does not exclude coincidences with other beta rays.

The electron spectrum coincident with the 600 kev gamma ray peak (Fig. 11) appears to be a single component, with an end point energy of 655 kev, agreeing with the value found by Cork _et al._[6] and by Keister, Lee, and Schmidt[8] for one component of the beta spectrum. The high energy end of the spectrum is obscured by a conversion electron line belonging to a gamma ray of about 600 kev energy.

The electron spectrum in coincidence with the 800 kev (Fig. 12) gamma rays has as its principal component a 655 kev beta ray, again with the high energy end obscured by a conversion electron line due to a 600 kev gamma ray. The low energy end does not drop off, as it does for the spectrum coincident with the 600 kev gamma ray, indicating that there may be electrons present from a component with end point at about 200 kev. In fact, when the distribution coincident with the 600 kev gamma ray is normalized to that for the 800 kev gamma ray, and subtracted, on the assumption that the 655 kev spectrum should

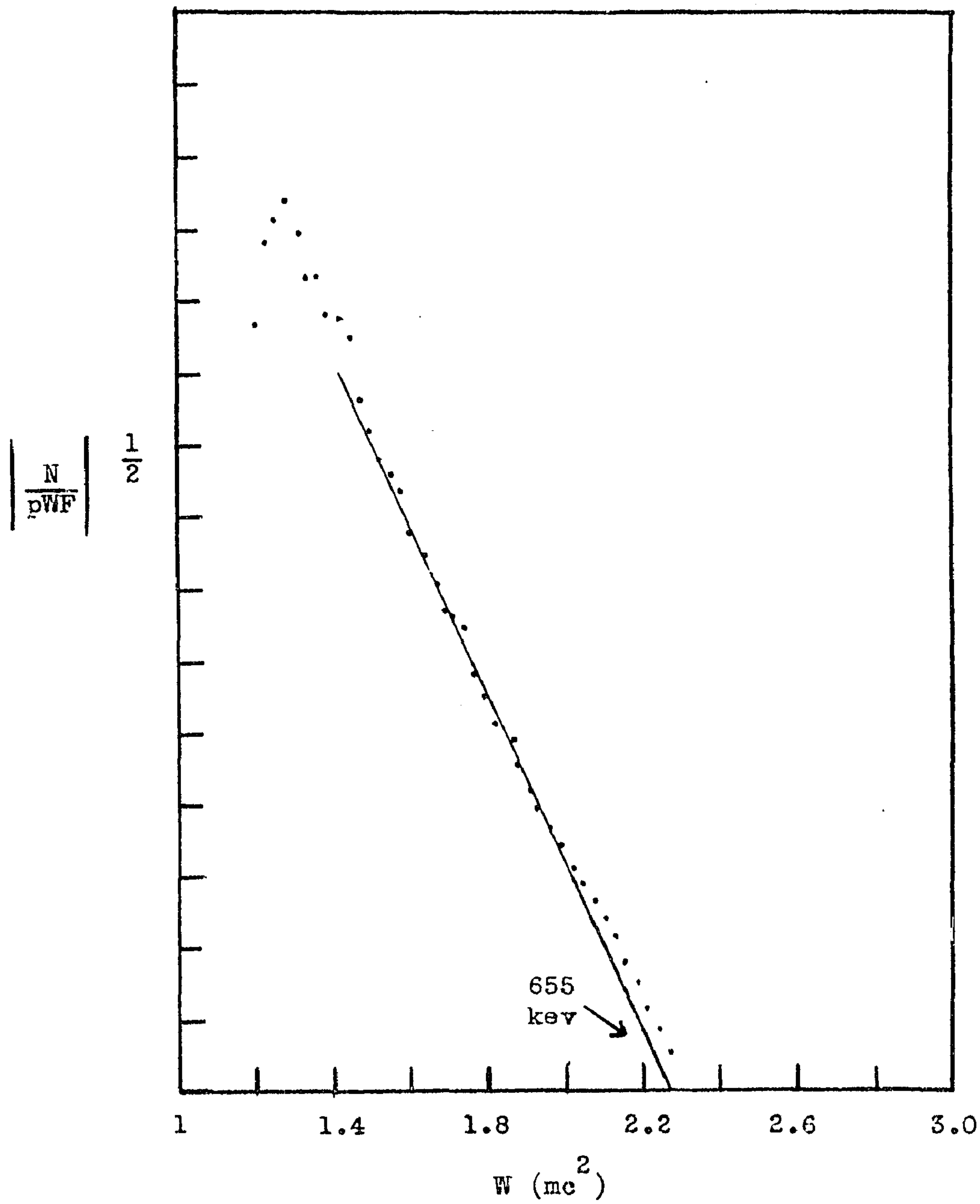

Figure 11. Fermi plot of the beta rays coincident with the 600 kev gamma rays.

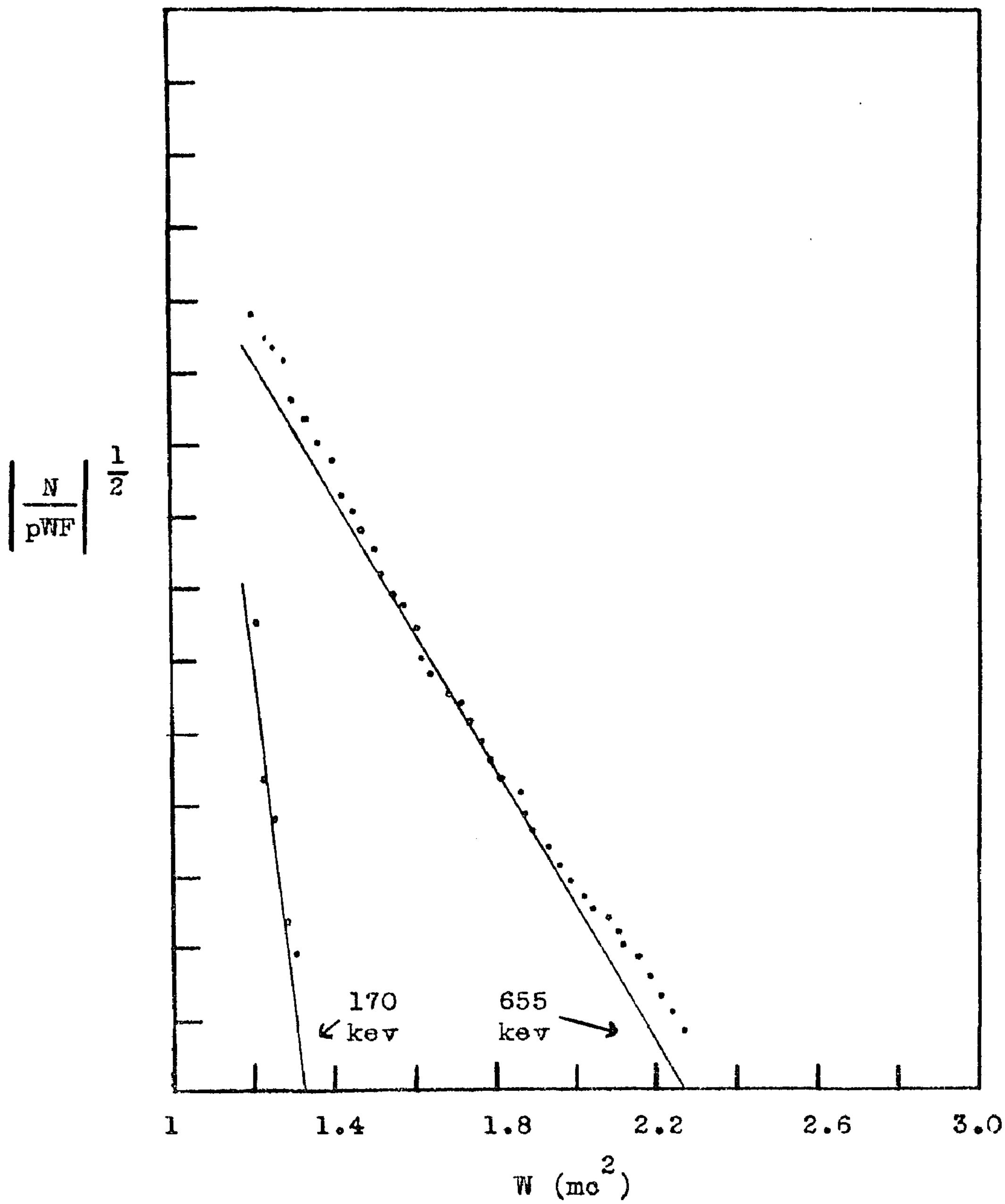

Figure 12. Fermi plot of the beta rays coincident with the 800 kev gamma rays.

have the same shape in both cases, the remainder is a component with an end point at approximately 170 kev. This is not consistent with the decay scheme proposed in the next chapter unless low energy transitions are allowed between levels not in the scheme leading ultimately to the 1401 kev level.

The electron spectrum coincident with the 1038 kev gamma ray must be corrected by subtracting 1/3 of the spectrum coincident with the 1367 kev gamma ray. The statistical errors inherent in the two sets of data are large, and such a procedure leads to a poor Fermi plot. This analysis showed a high energy component with an end point energy between 600 and 700 kev, and a low energy component between 200 and 300 kev. The high energy component may be identified with either the 655 kev component, or the 683 kev component of Keister, Lee, and Schmidt,[8] without conflict with this data.

The electron spectrum coincident with the 1168 kev gamma ray was corrected by subtracting 1/6 of the beta spectrum coincident with the 1367 kev gamma ray. Again the statistical errors limited the accuracy of the Fermi plot. The analysis yielded a complex spectrum with a high energy component between 600 and 700 kev, and a low energy component between 200 and 300 kev.

The electron spectron coincident with the 1367 kev gamma ray (Fig. 13) is complex, with a high energy component between 650 and 700 kev. It is not possible to decide whether these electrons are due to a 655 kev or a 683 kev beta ray, or both, because of the presence of two conversion electron lines. The conversion electron energies are such that the gamma ray energies would be 470 kev and 600 kev for K conversion.

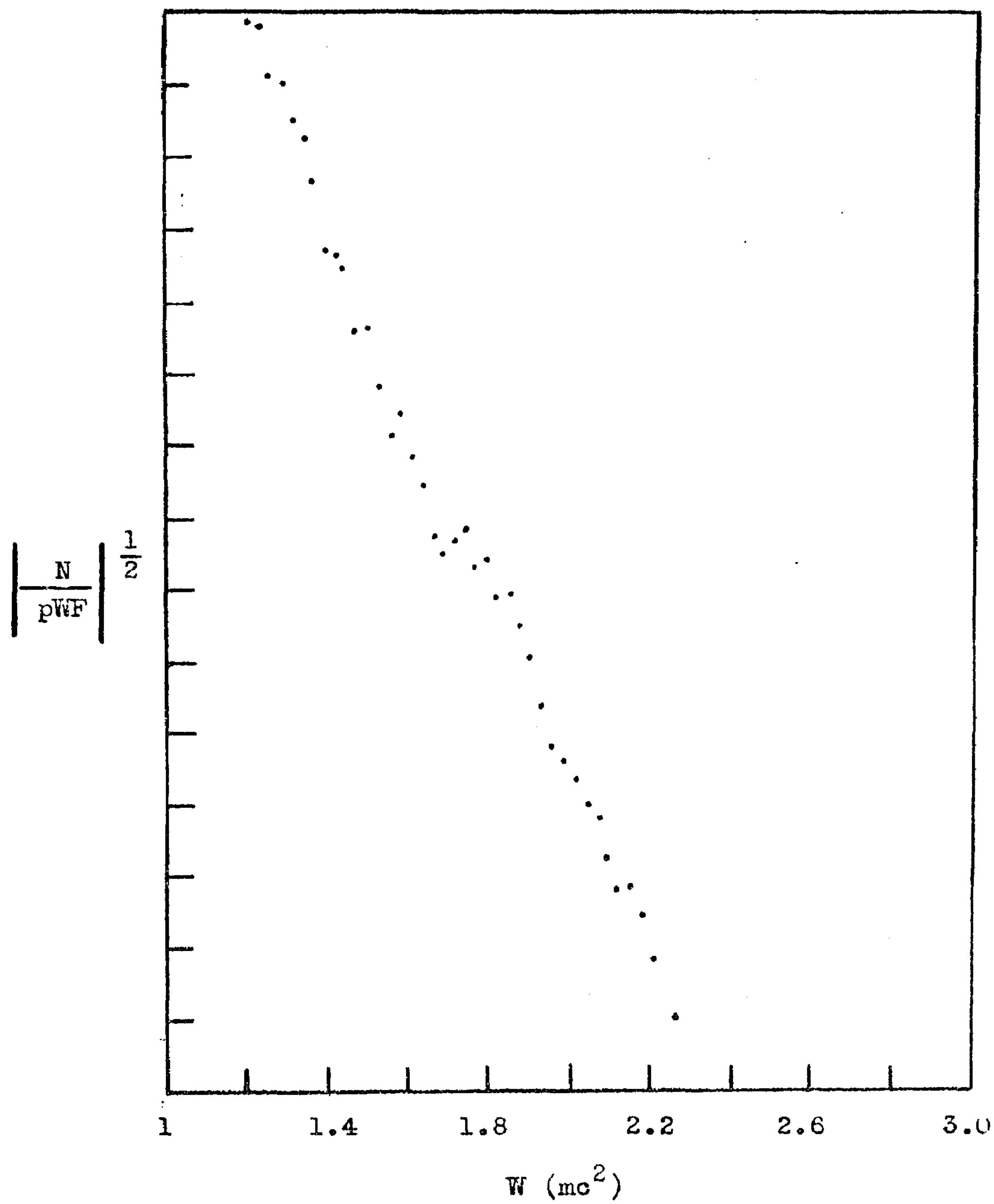

Figure 13. Fermi plot of the beta rays coincident with the 1367 kev gamma ray.

This provides evidence for the cascade of the 473 kev and 1367 kev gamma rays. The low energy component of the beta spectrum coincident with the 1367 kev gamma ray has an end point between 200 and 300 kev.

The electron component at 655 kev is required to satisfy the observed coincidences with the 600 and 800 kev gamma rays. While the existence of the 683 kev beta ray is not inconsistent with the data for the higher energy gamma rays, it is not definitely required by the data as is the 655 kev gamma ray. The decay scheme of the next chapter will be found to have no place for the 683 kev beta ray with the large (13%) percentage reported by Keister, Lee, and Schmidt,[8] and it is therefore preferred to identify the high energy beta components with the 655 kev beta ray.

The energies of the beta rays found in coincidence with the major gamma rays are collected in Table III, along with the fraction of the gamma ray decays which are coincident with the observed electrons. This fraction was calculated as in the case of the gamma-gamma coincidences. The sole difference is that the total electrons in coincidence are included, so $F = 1$. Also, the efficiency (corresponding to e_p) is 1.

No attempt was made to analyze the gamma ray singles spectrum. This has been done by A. N. Gabro.[11] The beta ray singles spectrum (Fig. 14) was taken with considerable care, in order to investigate the spectrum above the end point for the reported 655 kev and 683 kev components.

The work of August provided evidence for still another

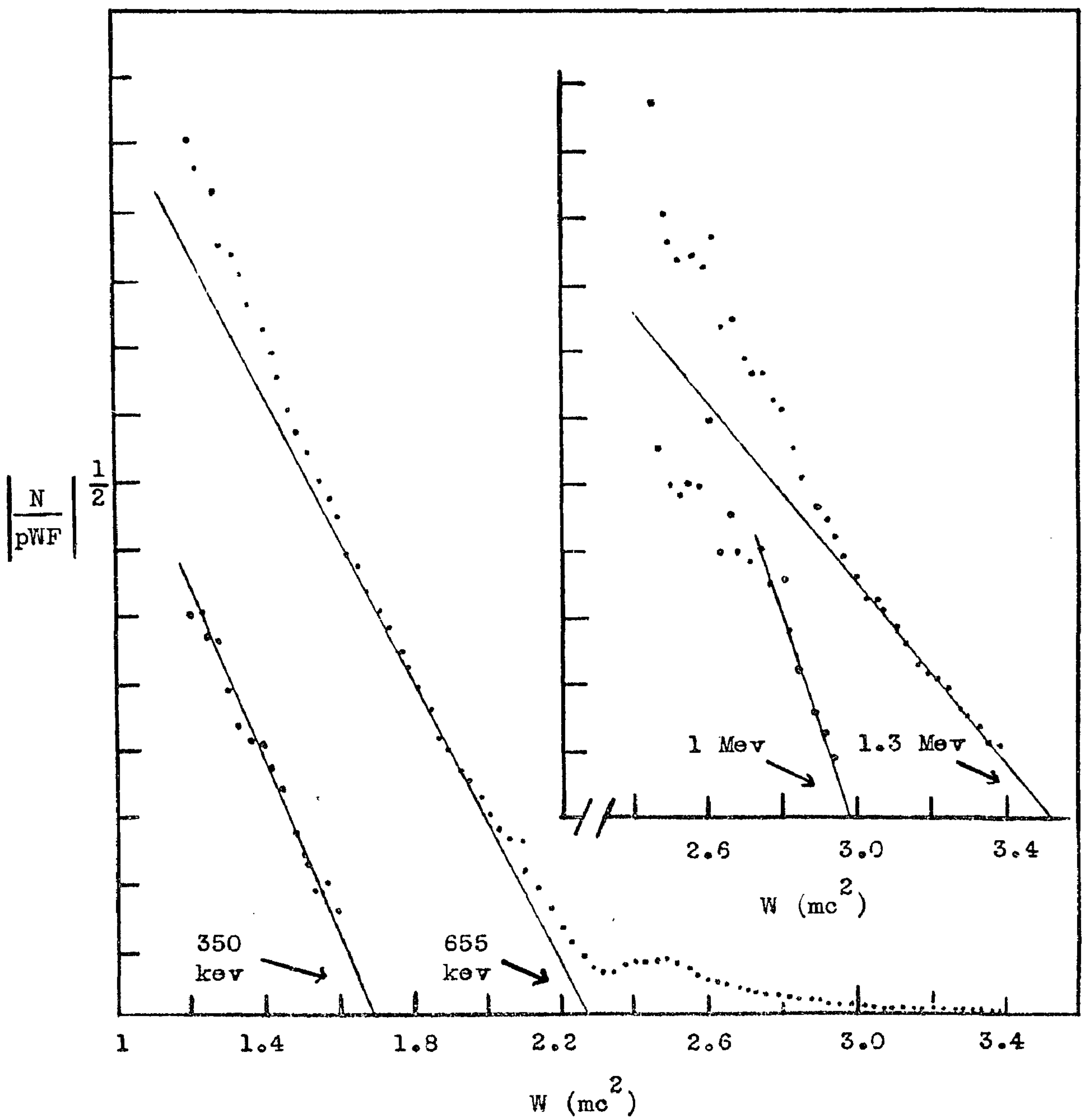

Figure 14. Fermi plot of the singles beta ray spectrum. The insert shows on an expanded scale the high energy portion of the spectrum after subtraction of conversion electrons.

TABLE III

BETA-GAMMA COINCIDENCES OBSERVED IN THIS WORK

Gamma ray energy (kev)	Beta rays in coincidence
473	200-300 kev (very few)
600	655 kev, 54% (total including conversion electrons)
800	200 kev, 655 kev, 77% (total including conversion electrons)
1038	200-300, 600-700, 24% (total including conversion electrons)
1168	200-300, 600-700, 7% (total including conversion electrons)
1367	200-300, 600-700, 38% (total including conversion electrons)

component with an end point energy of 1 Mev. This component is a very weak one, and it was hoped that the sensitivity of the scintillation spectrometer would be helpful in the investigation of it. The beta spectrum was followed over an intensity change by a factor of 10^5. The spectrum beyond the 655 kev component end point is complicated by the presence of conversion lines, due to the 800 kev gamma rays, the 1038, 1168 and 1367 kev gamma rays. These lines appear as broad peaks in the scintillation spectrometer spectrum.

Beyond the 655 kev component end point energy, the spectrum shows one or more components. After subtracting appropriate gaussian distributions for each of the 800 kev, 1038 kev, and 1168 kev conversion lines, the Fermi plot of the spectrum exhibits at least two distinct slopes. The subtraction of the conversion lines is subject to some inaccuracy. The relative intensities were determined from the data of Keister, Lee, and Schmidt,[8] and normalized to the visible peak, at an energy which allowed interpretation as the K conversion peak for the 796 kev gamma ray, and to the visible K conversion peak of the 1367 kev gamma ray. After subtraction, the Fermi plot at the position of the conversion lines for the 1038 kev and 1168 kev gamma rays showed small bumps, indicating that the subtraction process was not entirely accurate. In any event, if we accept Keister, Lee, and Schmidt's measurement of the relative intensities of the K conversion lines for these peaks, we find that they are an order of magnitude below the counting rates observed at the corresponding energies. The calculation of random sum pulses which would appear as pulses beyond the end point of the principal beta rays also yields a result an order of magnitude too low to account for the observations. This calculation was made by numerical integration of the observed spectrum over all possible sums. The existence of at least two components with energies above those reported by Cork _et al._,[6] and by Keister, Lee, and Schmidt[8] appears to be required by the data.

One of the new beta rays has an end point energy very close to 1 Mev, and may be the component observed by August. The other component has an energy in the neighborhood of 1.3 Mev.

Subtraction of a component with an energy of 655 kev from the principal part of the spectrum leaves what appears to be one lower energy component with an energy of 350 kev. The well known component at about 80 kev could not be observed in the experiment, due to the rejection of all energies below 90 kev in the beta spectrum.

In addition to the coincidences reported above, triple coincidences were observed between the 605 kev gamma ray, the 800 kev gamma rays, and a peak due to gamma rays of energy approximately 569 kev. In order to observe these coincidences, three NAI(Tl) crystals were employed, two 3" x 3" crystals, and one 1 1/2" x 1" crystal.

Two single channel analyzers were employed to select pulses in the photopeak for the 800 kev and 605 kev gamma rays. The 800 kev gamma rays could not be resolved. However, the pulse height selection for the 605 kev gamma ray was accomplished so that very few of either the 563 or 569 kev gamma rays were included in the other single channel. The fast-slow coincidence circuit was employed to gate the twenty channel analyzer whenever there was a 800-605 coincidence. A peak at 569 kev was observed in the twenty channel analyzer. It can not be decided whether any 563 kev gamma rays contribute to this peak, because the statistical errors are too large to permit a determination of the peak shape.

In addition to the beta-gamma coincidences noted above, other beta-gamma coincidences were obtained with the beta rays counted in the single channel analyzer instead of in the twenty channel analyzer. In this case, all betas above the 800 kev conversion electron peak were accepted in the single channel.

CHAPTER IV

THE DECAY SCHEME

A decay scheme consistent with most of the coincidence data is shown in Figure 15. The absence of any positrons, or of any Auger electrons due to a K capture branch, has lead previous investigators to assume that all the gamma rays observed are in Ba^{134}. Strong evidence for this assumption, as well as evidence against gamma rays due to impurities, lies in the fact that every gamma ray seen has been seen in coincidence with at least two other gamma rays. This leads to the conclusion that they are all in the nucleus Ba^{134}.

In order to see the reasons for proposing the decay scheme of Figure 15, let us first consider the beta-gamma coincidences. All of the higher energy gamma ray peaks, 600 kev and above, showed coincidences with beta rays between 600 and 700 kev; in particular, the 600 kev and 800 kev peaks were found to be in coincidence with the 655 kev beta ray, while in the case of the others the beta ray end point could not be given so definitely. In these cases it is reasonable to assume that the beta ray is the 655 kev, or perhaps the 683 kev beta ray reported by Keister, Lee, and Schmidt.[8] In any event, these gamma rays must arise from levels as low as, or lower than, the level to which the 655 kev beta ray leads.

The fact that the two strongest gamma rays, namely the

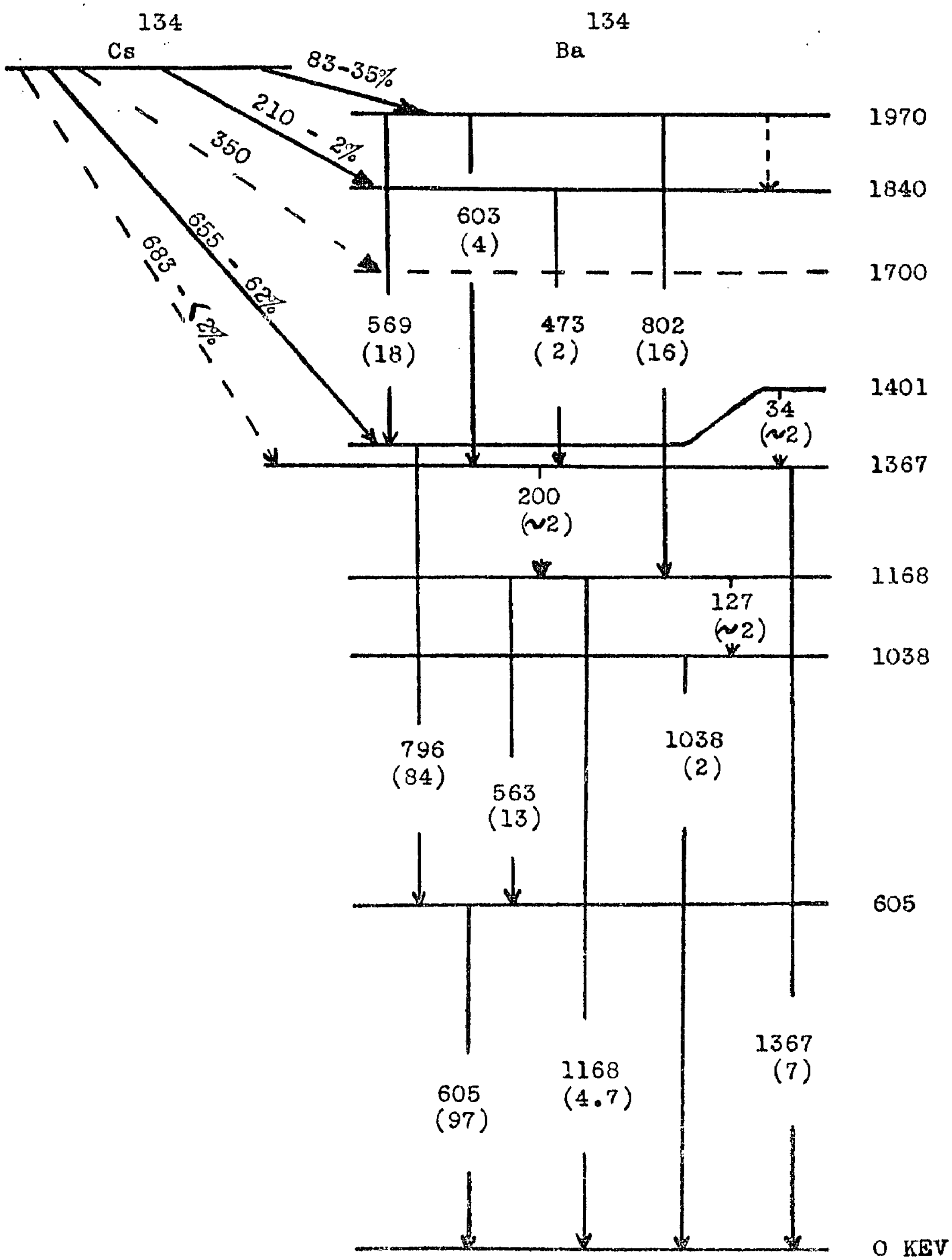

Figure 15. Decay scheme proposed in this work.

790 kev and 605 kev, are in approximately 100% coincidence implies that they are in sequence, and that the lower one goes directly to the ground state. This establishes a level in Ba^{134} at 1401 kev. Furthermore, the high coincidence between the 655 kev beta ray and each of these gamma rays implies that the 655 kev beta ray goes directly to the 1401 kev level. This gives a total energy of 2056 kev for the decay, and agrees with previous work.

The fact that the 605 kev gamma ray is slightly the stronger of the two implies that the 796 kev gamma ray is first in the sequence. This is further supported, as will be seen in the next paragraph, by the coincidences between the 605 kev gamma ray and another at very nearly this same energy. A 605 kev level is thus established.

Consideration of the 1367 kev gamma ray data shows that it must be in sequence with a 600 kev gamma ray, but also that it must be coincident with 655 kev (or higher) beta rays. The latter fact shows that the 1367 kev gamma ray must follow the 600 kev gamma ray. Since we have already allowed a 605 kev gamma ray to go to the ground state, we must assume that there is another 600 kev gamma ray. Such a gamma ray has been postulated by Keister, Lee, and Schmidt[8] to account for their data. The energy sum for a 603 kev gamma ray and the 1367 kev gamma ray is 1970 kev. It is therefore postulated that there is a 1970 kev level with a 603 kev gamma ray going to a 1367 kev level, and that a 1367 kev gamma ray goes to the ground state from there. This will account for the 600-600 gamma-gamma coincidences provided the 796-605 kev sequence is in the same order given in the last paragraph.

Coincidences between the 800 kev peak and the 1168 kev gamma ray require that there be a sequence involving an 800 kev and a 1168 kev gamma ray. The sum of the energies is 1968 kev, and this suggests that the sequence starts at the 1970 kev level and is fed by the generally accepted beta ray of 80-85 kev. The beta-gamma coincidences for the 1168 kev gamma ray show that it is the one of this pair which goes to the ground state. These results require that we have a 1168 kev level and a second 800 kev gamma ray which is to be identified with the 802 kev gamma ray reported by others.

That the 1038 kev gamma ray arises from some level below the 1401 kev level is required by the beta-gamma coincidences. One can be convinced then, that the 1038 kev gamma ray either has to go to the ground state, (if we assume a total energy difference between Cs^{134} and Ba^{134} of the order of 2050 kev, in accord with the assumption of 1970 kev as the highest gamma ray level), or be followed by a low energy gamma ray of 200-400 kev.

The latter alternative being less attractive, the 1038 kev gamma ray is assumed to go to the ground state. This requires a 1038 kev level.

The above considerations account for 6 of the 9 gamma rays recorded by Keister, Lee, and Schmidt[8]. The 473 kev gamma ray can be accounted for with some confidence if we assume that there is a 210 kev beta ray. This is in accord with the assumption of a 215 kev beta ray by Keister, Lee, and Schmidt[8] and is consistent with the resolution of the beta spectrum by Cork *et al.*[6] There would then be an 1840 kev level, from which the 473 kev gamma ray could proceed to the 1367 kev level. That the 473 kev and 1367 kev gamma rays are in coincidence is shown by the 440 kev conversion electrons superimposed on the 1367 kev beta coincidence data.

The 569 kev gamma ray is assumed to be a transition between the 1970 and the 1401 kev levels. The coincidences observed for the 600, 800, 1038, 1167, and 1367 kev gamma ray peaks show a photopeak between 560 and 570 kev. This implies that one (or both) of the 563 and 569 kev gamma rays occurs above the 1401 level. Since the energy difference between the 1970 and 1401 levels is 569 kev, it is assumed that the 569 kev gamma ray is to be placed there. This will account for the observed coincidences. The 796-605-569 kev triple coincidences observed lend support to the above assignment of the 569 kev transition.

Of the nine principal gamma rays, only the 563 kev gamma ray is still to be accounted for. Keister, Lee, and Schmidt[8] observed that "very few if any" of the 563 kev gamma rays are in coincidence with high energy beta rays (by which they seem to mean electrons above 90 kev). However, intensity data obtained in this experiment suggest that a 563 transition from the 1168 kev level to the 605 kev level may occur. The 1168 kev level is populated so strongly--principally by the 802 kev gamma ray--that another transition from the 1168 kev level is needed. The intensity of such a transition to account for the 1168 kev level population is of the same order as the intensity of the 563 kev gamma ray as found by Gabro[11] and Keister et al.[8]

There occurred in the coincidence spectrum of each of the 473, 1038, and 1167 kev gamma rays, and less certainly with the 1367 kev gamma ray, a peak at 280 kev. The width of this peak is in all cases greater than a gaussian distribution at this energy should be. It is suggestive that some of this peak may be due to gamma rays scattered from some fairly localized mass in the vicinity. The most appropriate supposition would be that the scattered photons were Compton photons due to scattering of the 600 and 800 kev gamma rays, in which case the scattering angle would be of the order of 90°. The lead shield which was used to prevent scattering from one crystal to the other was in a position to cause such scattering. Even when back scatter is allowed for, the peak appears to be too broad to be due to a single gamma ray, and so one must account for two or more gamma rays in the vicinity of 280 kev. It is not clear what role such gamma rays play in the decay scheme.

In order to explain coincidences between the higher energy gamma rays and a gamma ray of the order of 560 kev, it is assumed that there is a 34 kev transition between the 1401 and 1367 kev levels. This transition would proceed almost entirely by L conversion, and the electrons would probably not be seen, since the transition would be very weak due to the competition of the 796 kev gamma ray. A 200 kev transition is assumed between the 1367 and 1168 kev levels, and a 127 kev transition between the 1168 and 1038 kev levels. There is some suggestion of these in the coincidence data. There is also the suggestion of a 130 kev gamma ray in coincidence with the 473 kev gamma ray. This transition is possibly one between the 1970 and 1840 kev levels.

The analysis of the beta spectrum, as previously mentioned, showed two high energy components, above the 655 kev end point. The higher energy one of these beta rays requires that there be another level between the 605 kev and 1038 kev levels. The best determination of the end point of the beta ray gives 1300 $\pm$ 100 kev. This leads to a level above the 605 kev one, although the possibility that the beta ray leads to the 605 kev level cannot be excluded.

The low energy 350 $\pm$ 50 kev beta requires a level between the 1840 kev and the 1401 kev levels. The best determination of the beta ray end point energy gives 1700 kev as the level energy.

Coincidences obtained between the beta rays above 800 kev and the gamma ray spectrum show a broad peak at 210 kev, a peak of equal height at 605 kev, and a smaller peak at 800 kev. If the 800 kev peak is ascribed to coincidence with conversion electrons of the 1038 kev and 1168 kev gamma rays, nearly all of the 210 kev and 600 kev coincidences remain after subtraction of the proper fractions of the 1038 kev and 1168 kev coincidence distributions. Since the 1367 kev conversion electron total is not greatly different from that of the 1038 kev, or the 1168 kev gamma ray, the remaining 600 kev counts due to conversion electrons can be accounted for when a fraction of the 1367 coincidence distribution equal to that for the 1168 kev is subtracted. When this is done, one half of the 600 kev counts remain. Calculation of the fraction of the high energy beta rays which are followed by each of the 210 and 600 kev gamma rays shows that all of these beta rays are in coincidence with each peak. This indicates that all of the levels populated by high energy beta rays are de-excited by transitions leading through the 605 kev gamma ray. However, it is not possible to determine the exact energies of the levels fed directly by the high energy beta rays from the analysis of the beta spectrum. For these reasons, the exact level scheme cannot be determined in the region between the 605 kev and 1040 kev levels.

It is worthwhile to consider the intensities of the gamma rays and beta rays in connection with the decay scheme, to see if the qualitative arguments from observed coincidences are consistent with quantitative predictions of relative intensities, and with the observed coincidence fractions. In order to determine this, it is necessary to have the relative intensities of the gamma rays as observed.

Relative intensities have been assigned (numbers in parenthesis in Figure 15), using the values found by Gabro.[11] In the cases of the 563-569 kev and 796-802 kev pairs, the relative intensities assigned by Keister, Lee, and Schmidt[8] have been used for the pair, but the total intensity has been made to agree with Gabro's results in each case.

The low energy gamma rays have been assigned intensities with account being taken of population of the levels involved and of the coincidence fractions wherever applicable.

The upper 600 kev gamma ray has been assigned an intensity to agree with the observed coincidence fraction for the 1367 kev gamma ray.

The relative intensities assigned agree remarkably well with the transition rates required by the decay scheme. The coincidence fractions to be expected on the basis of the relative intensities and the decay scheme are in agreement within the limits of error with the observed coincidence fractions, except perhaps for some of the low energy gamma rays. These will in general be more highly converted than the higher energy gamma rays, and this may account for the low fractions observed in these cases.

The decay scheme proposed agrees exceptionally well with the experimental findings. The scheme was proposed at first to account qualitatively for the observed coincidences. The consistency which the scheme shows in accounting for the observed intensities is then strong evidence in its favor. A full investigation of the role played by the low energy gamma rays will probably require techniques which are not available at the present time.

The high energy beta rays cannot be assigned an exact energy, but the existence of strong coincidences between them (of the order of 100%) and the 605 kev gamma ray leads to the conclusion that while there must be one or more levels not shown in the scheme in the region between the 605 and 1038 kev levels, these do not lead to any new gamma rays of appreciable intensity leading to the ground state.

The 350 kev beta ray which was found from analysis of the beta spectrum leads to the inclusion of a 1700 kev level. Unless this level is the origin of the gamma rays of the order of 280 kev observed in coincidence with the high energy gamma rays, there is no place for such a level in the decay scheme. Because of the large uncertainty in energy for the beta ray, it is possible that the level should be of the order of 280 kev above the 1401 level, or at 1681 kev.

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VITA

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